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<b>(54) Title:</b> THERMALLY CONDUCTIVE FILM AND METHOD FOR THE PREPARATION THEREOF  <b>(57) Abstract</b>  Thermally conductive interface materials comprise a matrix resin film filled with carbon fiber oriented transversely to the plane of the film and extending substantially through the film thickness to provide a high conductivity path between the adhered components. The interface materials may be particularly useful as thermally conductive adhesive films for bonding electronic components to the surfaces of heat sinks and thermal management devices.		

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THERMALLY CONDUCTIVE FILM  
AND METHOD FOR THE PREPARATION THEREOF

BACKGROUND OF THE INVENTION

5 This invention relates to thermally conductive film adapted for use as an interface material in a variety of thermal management applications. More particularly, the invention relates to thermally conductive thin film having very high thermal conductivity in the through-thickness direction, i.e. along the z axis of the film, that may be found particularly useful as an interface material, and as a thermally conductive adhesive or sealant in thermal management devices.

10 The invention may further be described as relating to methods for the production of such film.

Heat generation is a problem in a great variety of electronic devices comprised of a semiconductor component such as, for example, laser diodes, light-emitting diodes, thyristors, microwave electron transfer devices and the like. Semiconductor or multichip modules that consist, for example, of monocrystalline silicon, wherein millions of transistors are positioned  
15 on or near a single surface of the chip, produce considerable amounts of heat energy in operation. Removal of byproduct heat is important to the lifetime of electronic components, and the art has continually sought improved means for managing the high levels of heat output associated with such devices.

Early thermal management practice in the design of carriers, heat sinks and housings relied on  
20 the use of metals and alloys selected for their high thermal conductivity. More recent innovations and modifications in materials have included, for example, combinations of metal housings with metal-coated diamond chips or wafers, intended to take advantage of the fact that diamonds have the highest thermal conductivity known, and metal matrix composites filled with diamond particles. Other solutions for thermal management problems, particularly  
25 designed for use with high power density devices, include liquid-cooled heat sink structures and chip module housings that rely on liquid nitrogen as the coolant.

Removal of heat from high density multichip modules generally takes place at the module's backside, i.e. the surface of the chip opposite the surface where the transistors are positioned. To maximize the heat transfer the module is preferably secured in direct contact with a heat  
30 sink. Other design approaches include interposing conductive thermal planes to transfer heat

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from the electronic components to a heat sink. The thermal planes, conventionally aluminum or copper members, are surface-mated to electronic devices or associated printed circuit boards and interfaced to the heat sink at the edges of the thermal plane.

The method of attaching the components to the heat sink or thermal plane may greatly affect the thermal efficiency of the device. Direct, rigid attachment is often not a desirable solution. A mismatch between the coefficients of thermal expansion (CTE) of the semiconductor device and the heat sink device will, as a consequence of the heat produced during operation, subject the brittle semiconductor to mechanical stress, the level of the stress imposed on the device being a function of the degree of mismatch. If the stresses are severe and uncompensated, catastrophic failure can occur, shortening the service life of the module. Similar problems arise in most situations where heat generation sufficient to damage the device can occur.

Air is a good thermal insulator, and closing the airgap formed at the interface of a component held in close surface contact with a thermal plane or heat sink is thus very important for efficient thermal management. Conventionally, airgaps may be closed by use of thermal interface materials such as, for example, thermally conductive greases or deformable heat conductive pads, in combination with metal clips or the like designed to hold the parts tightly together. Thermally conductive adhesives in the form of liquids or tapes are also known, and these are widely used to bond components and heatsinks while serving as a thermally conductive filler. Such adhesives are also widely used for bonding chip carrier structures, substrates, heat sinks and the like, and for adhering the die or chip, for lid sealing and the like.

Organic-based adhesives are typically filled with particulate metal fillers or with inorganic fillers such as alumina to provide thermal conductivity. Metal foils surface-coated with a suitable adhesive are also employed for these purposes. Thermally conductive adhesives are preferably tailored in their CTE characteristics through selection of filler and matrix components, thereby facilitating a match in CTE between the components. Adhesives may be used in paste form or, more conveniently, applied as a thin film and then bonded, usually by application of heat. These and a variety of related adhesive materials for electronic applications are well known in the art.

The addition of conductive fillers to resins is thus well known and widely used as a method for improving dimensional stability and to increase the thermal conductivity of the resin. However, the fillers and methods that have been previously disclosed in the art have only

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afforded modest increases in thermal conductivity, generally no greater than 20 times that of the base resins. More commonly, the thermal conductivity for filled thermoset resins used commercially in the electronics industry have been on the order of 2 to 4 W/mK, and injection moldable filled thermoplastic formulations are available having thermal conductivities in the 4 to 9 W/mK range.

Resins filled with uniaxially oriented thermally conductive fiber may vary in thermal conductivity, exhibiting high conductivity along the fiber direction and low conductivity when measured transversely to the fiber direction. For example, for a structure comprising aligned continuous carbon fiber embedded in a film, the thermal conductivity along the fiber axis will be determined by the "rule of mixtures" and thus equal the area of fiber times the fiber conductivity plus the area of resin times the resin conductivity. Measured in the out-of-plane direction, i.e. along an axis perpendicular to the fibers, the thermal conductivity is dominated by the resin, and therefore found to be considerably less. The case of resins filled with discontinuous fiber is more complex; however, it may generally be said that the thermal conductivity measured along any axis will be strongly dependent upon the fraction of fibers oriented along that same axis.

Filled molding resins and molded articles filled with thermally conductive filaments such as pitch-based graphite fiber were more recently disclosed in the art. The fiber component of these materials is discontinuous and said to be distributed through the structure in a three-dimensional arrangement. An essentially three-dimensional isotropic distribution of reinforcing fiber was obtained and found to significantly improve the dimensional stability of the parts. However, the thermal conductivity reported for these filled materials was only about 5-9 W/mK, a very modest improvement over filled thermoplastics disclosed in the prior art. Though not disclosed particularly for use as thermally conductive adhesive, carbon fiber-filled thermoplastic resins containing from 10 to 60 wt% carbon fiber are also widely known in the art. Thermoset compositions comprising from 20-60 wt% carbon fiber have also been described for use as adhesives in electronic applications, for example in US 5,026,748.

The filled resin formulations presently available for use as adhesive materials in electronic applications are thus generally low in thermal conductivity. Even the most conductive has a thermal conductivity below 10 W/mK, while most are much lower, generally as low as 2 to 3

W/mK. Adhesive resin compositions with thermal conductivity from 50 W/mK to as great as 100 W/mK and greater appear to be unknown in the art.

The electronic arts thus continue to lack adhesive materials having thermal conductivities adequate to meet the thermal management considerations imposed by current trends in electronic packaging. Thermal conductivity is becoming an increasingly important design consideration, and a ten- to one hundred-fold increase in thermal conductivity, particularly in the through-thickness direction, for thermally conductive adhesives would thus represent a substantial advance in the art. Adhesive film structures exhibiting these highly desired thermal properties and a method for making such structures would be particularly useful for meeting the growing demand for more highly conductive thermal management materials.

### SUMMARY OF THE INVENTION

The highly thermally conductive adhesives of this invention comprise a resin selected from thermoplastic resins and thermoset resins together with pitch-based carbon fiber. The formulations and films according to the invention have a thermal conductivity greater than about 100 W/mK or more, and are particularly well-suited for use as thermally conductive adhesives in bonding components of devices where thermal management is an important requirement. The method employed for fabrication of these adhesives in film form is readily applicable to providing thermally conductive film suitable for use in a wide variety of electronic applications.

### DETAILED DESCRIPTION

The thermally conductive film and film adhesives of this invention will comprise carbon fiber embedded in a polymeric matrix material such as, for example, a thermoplastic resin or a thermoset resin composition. The carbon fiber will be aligned and oriented transversely to extend between the faces of the film and provide a continuous high conductivity path between the adhered components.

Carbon fibers suitable in the practice of this invention include highly graphitized carbon fiber having high thermal conductivity and a low or negative coefficient of thermal expansion produced from pitch. As used herein, the term "carbon fibers" is intended to include graphitized, partially graphitized and ungraphitized carbon reinforcing fibers or a mixture

thereof. The preferred carbon fibers will be pitch-based carbon fiber. Such fiber generally will have a thermal conductivity greater than about 300 W/mK. The carbon fiber will preferably have a thermal conductivity greater than about 600 W/mK. Such fiber having a conductivity greater than 900 W/mK, more preferably above 1000 W/mK, will be particularly preferred.

5 Fiber with even greater thermal conductivity, from as high as 1300 W/mK up to the thermal conductivity of single crystal graphite, 1800 W/mK and higher, will also be suitable. Although lower conductivity fiber, for example PAN-based carbon fiber, typically having a thermal conductivity in a range of from 10-20 W/mK, may also be employed in manufacturing thermally conductive compositions according to the processes employed in the practice of this invention, the transverse thermal conductivity of the resulting film adhesive would be  
10 undesirably low, even with very high fiber loading. Thus, such fiber will not be preferred.

Pitch-based carbon fiber in a variety of strengths and conductivities is readily available from commercial sources. Fiber with thermal conductivity falling in the range of from 600 W/mK to greater than 1100 W/mK, a density of from 2.16 to above 2.2 g/cc and a very high tensile  
15 modulus, from  $110 \times 10^6$  psi to greater than  $120 \times 10^6$  psi, is readily obtainable from commercial sources, including Thorne<sup>®</sup> pitch-based carbon fiber from Amoco Polymers, Inc. Commercial carbon fiber is ordinarily supplied in the form of continuous carbon fiber tow or yarn comprising a plurality, usually from 1000 to 20,000 or more, of carbon filaments 5 to 20  
20 microns in diameter with the axially-aligned filaments providing strength in the fiber direction of the tow.

The matrix resin component of the invented film adhesive compositions may comprise a thermoset resin or a thermoplastic resin, and mixtures of such resins may also be found useful for these purposes. Generally, thermoset resins will be preferred for use where resistance to flow and to distortion under mechanical and thermal stress are important considerations.  
25 Conventional thermoset resins that may be found suitable for this use include epoxy resins, cyanate resins, bismaleimide resins, thermosetting phenolic resins including novolacs, resoles and the like, and thermoset polyesters. These and similar thermoset resins are widely employed in the adhesives, laminating and coatings arts, and many such resins will be found suitable for this use.

30 Elastomers, including silicone, urethane, acrylic and similar elastomeric resins that are widely employed as adhesives and sealants, may also be found useful in the practice of this invention.

Many such elastomers, when fully cured, have the physical properties needed to withstand the wide range of thermal and environmental conditions encountered in electronic devices. Such materials are readily formulated according to practices well understood in the elastomer arts to provide wide range of soft and readily deformable interface materials, and alternatively, materials that are hard and rigid, and thereby less subject to creep under mechanical stress.

Ultrahigh melting temperature thermoplastic resins such as aromatic polyimide resins, polyamide-imide resins, polyphenylene resins and the like, many of which have decomposition temperatures that lie near or even below their softening or thermal processing temperatures, may also be useful in combination with carbon fiber tow to provide film adhesive materials.

Although these resins are difficult to mold and lack the ease of fabrication which characterizes engineering thermoplastics, they exhibit good dimensional stability under load and excellent thermal and chemical resistance, and are highly desirable for use in demanding environments.

Extrudable and injection moldable resins generally known in the art, including the high performance thermoplastics widely known as engineering thermoplastics, may also be found suitable for this use. Among such resins are the aliphatic polyamides including nylon 6, nylon 6,6, nylon 4,6, nylon 11 and the like; polyphthalamides, including polymers of one or more aliphatic diamines such as hexamethylene diamine, 2-methylpentamethylene diamine and the like with terephthalic acid compounds; copolymers thereof with additional dicarboxylic acid compounds such as isophthalic acid, adipic acid, naphthalene dicarboxylic acid and the like; polyarylate resins including polyethylene terephthalate (PET) resins, polybutylene terephthalate (PBT) resins and the like; arylene polycarbonate resins including poly(bisphenol A carbonate); the well known polyaryl ether resins such as PPO resins, including the thioether analogs thereof such as PPS resins and the like and the corresponding sulfone- and ketone-linked polyaryl ethers such as polyether sulfones, polyphenylether sulfones, polyether ketones, polyphenyl ether ketones and the like, many of which are widely available from commercial sources; and the variety of aromatic polymer resins widely known and sold as liquid crystal polymer (LCP) resins. A great many such thermoplastic resins are commercially available from a variety of sources. As will be readily understood, where use of a thermoplastic resin matrix is contemplated, the thermoplastic resin will be selected to provide an interface material having flow and melt temperature characteristics appropriate to the thermal conditions that will be encountered in use.



Generally, the film adhesive resin formulations useful as interface materials according to the invention will comprise from about 10 to about 70 vol%, more preferably from about 30 to about 60 vol% carbon fiber and, correspondingly, from about 10 to about 30 vol%, more preferably from about 70 to about 40 vol% of the resin component. Alternatively, the  
5 formulations may be conveniently described as comprising from about 15 to about 80 wt%, preferably from about 15 to about 70 wt%, still more preferably from about 25 to about 70 wt% carbon fiber.

When appropriately formed as adhesive film as described herein, applied to the component articles and, in the case of thermoset resins and elastomers, thermoset or cured, the filled resin  
10 film adhesives and sealants of this invention form bond layers with extraordinarily high thermal conductivity, as great as 50 W/mK or more, together with excellent dimensional stability over a wide range of temperatures.

The film adhesive of this invention will be constructed to provide a high degree of thermal conductivity when measured in the through-thickness direction. The high thermal conductivity  
15 will be accomplished by having carbon fibers extending substantially through the film thickness, thereby providing a continuous thermal and electrical path between the opposing faces of the film. Substantially all of the fiber component will preferably be aligned in the through-thickness direction, and most preferably will be aligned perpendicular to the film plane. It will be readily apparent that fiber extending through the film thickness and inclined at  
20 an angle, rather than being normal to the surfaces of the film, will also provide the necessary conductive path.

The thermal conductivity of the film in the through-thickness direction will be very dependent on the amount of fiber extending between the opposing surfaces. In order to obtain a high through-thickness thermal conductivity, the proportion of carbon fiber component oriented  
25 substantially in the plane of the film will thus be low, preferably amounting to less than 50%, more preferably less than 25% of the total volume of the carbon fiber present in the film.

The thickness of the film adhesive will depend in part upon the intended use. For bonding and adhesive applications the general and preferred practice is to employ bonding layers having the least practical thickness. For such use the film thickness will generally lie in a range of from  
30 0.005 cm (2 mil) to about 0.25 cm (100 mil), preferably from about 0.005 cm to about 0.01 cm. For electrically conductive filler applications, particularly where gap-filling is a necessary

function of the filler, greater film thicknesses, as much as 0.25 cm, may be found useful. Although the thermal conductivity of the film will be quite good even when the thickness becomes vanishingly small, films having a thickness of less than about 0.005 cm will generally be found to be impractical for most adhesive uses.

- 5 The films of this invention may be conveniently fabricated by a variety of methods. In one embodiment, lengths of unidirectional prepreg tape, i.e. a carbon fiber prepreg in the form of a tape or ribbon having conductive fiber aligned along the longitudinal axis of the tape and impregnated with thermoplastic or uncured thermoset resin, will be plied or stacked to provide a thick, multi-layered body with substantially all of the fiber uniaxially aligned. The layered  
10 body or laminate will then be sliced into thin sheets by cutting along a plane substantially perpendicular to the fiber axis of the body, providing a wafer or film having substantially parallel faces, with fiber uniformly distributed and extending through the thickness of the slice.

- The slicing or cutting operation may be carried out by any of a variety of means, including use of diamond saws, rotary or vibratory microtomes, string saws, etc. The selection of the cutting  
15 device will depend in part on the nature of the carbon filled resin, particularly including deformability and hardness character. Generally, when the matrix resin is an uncured thermoset resin or a soft, elastomeric material, it may be preferable to chill the structure prior to slicing. Chilling may effectively stiffen the matrix, thereby reducing the likelihood of  
20 distorting the laminate and displacing fiber during the slicing operation. For thermoplastic matrix resins, particularly for high temperature resins and resins that are hard or that may tend to be brittle, it may be desirable to perform the slicing at an elevated temperature, thereby softening the matrix resin to improve processing and reduce brittleness.

- Other embodiments of methods suitable for forming a body comprising uniaxially aligned carbon fiber embedded in a resin matrix for slicing into film or wafers include, for example,  
25 gathering a plurality of prepreg tows comprising continuous carbon fiber impregnated with matrix resin to form a rod-like body or structure having the conductive fiber aligned along the axis. The resulting structure, substantially circular in cross-section as formed, may be used in that form or shaped using suitable tooling to provide structures having cross-sectional configurations as desired, including rectangular or square. Slicing perpendicular to the fiber  
30 axis, as described herein, will provide wafer or film with fiber extending through the thickness of the slice. In another process embodiment, tow or uniaxially aligned fiber may be first

collected and placed into a suitable mold cavity and then impregnated to provide a body comprising fiber embedded in a resin matrix for slicing into thin wafers or film as described herein above.

It will be further apparent that slicing perpendicular to the fiber axis may be done along one of the two orthogonal axes, and inclined at an angle as great as 45° from perpendicular along the remaining axis. Wafers or film obtained in this manner will have carbon fiber that extends through the thickness at an angle other than normal to the surfaces of the film.

It may be desirable for particular applications to further include in the matrix resin such plasticizers and processing aids, as well as thermal stabilizers, oxidation inhibitors, flame retardants, reinforcing fillers and fiber, dyes, pigments and the like as are conventionally employed in the compounding arts for use with adhesive resin materials. It will be readily apparent that the utility of the filled film materials of this invention lies in the substantial thermal conductivity exhibited by these materials and, where employed as adhesives, in the ability to adhere to the substrates that will be bonded. These additional components, as well as the amounts employed, will thus be selected to avoid or at least minimize any undesirable reduction in thermal conductivity or adhesive properties.

The formulation and compounding of thermally conductive interface materials of the invention will be better understood from a consideration of the following illustrative examples.

#### EXAMPLES

The test methods employed in the following examples for determining mechanical properties are described in ASTM D4018 and D3800.

Example 1 Unidirectional prepreg tape comprising aligned tows of high modulus, pitch-based carbon fiber was impregnated with a conventional cyanate ester-epoxy novolac thermosetting prepreg resin formulation. The resin was prepared substantially as described in Example 26 of U.S. 4,804,740.

The carbon fiber 2000 filament tow was obtained as Thornel® K-II00X pitch-based carbon fiber from Amoco Polymers, Inc. The published nominal properties for such fiber include thermal conductivity of 1050 W/mK, tensile modulus of 130 Mpsi, a tensile strength of 450 Kpsi, a density of 2.2 g/cc, and a CTE of -1.5 ppm/°C.

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The impregnated prepreg tape with a resin content of 35.3 wt% and areal weight of 262 g/m<sup>2</sup>, was cut into strips 1 cm in width by 3 cm in length. The strips were plied to provide an 11-layer laminate, which in turn was cut to a length of 1.5 cm and again plied to give a 1 cm x 1.5 cm laminate having a total of 22 unidirectionally-aligned fiber layers and a thickness of about 0.3 cm.

The laminate was then placed in a fixture and sliced perpendicularly to the fiber axis using a steel razor blade. The pieces of film adhesive thus obtained had transversely oriented carbon fiber embedded in a resin matrix and measured approximately 0.3 cm x 1.5 cm, having a thickness of 6-10 mils, or 0.015-0.025 cm.

The adhesive film strips were gold-coated and examined by scanning electron microscopy (SEM). The surfaces of the films were seen to consist of exposed ends of carbon fiber embedded in the resin matrix.

Example 2 Prepreg tape approximately 4.5 inches in width, prepared as in Example 1, was cut into nominal 1 inch lengths and plied to provide a 6-ply laminate. The laminate was then sectioned lengthwise (along the fiber direction) to give 6 strips 1 inch in length and measuring approximately 3/4 inch in width. These were then plied to have aligned fiber, giving a 36-ply laminate measuring approximately 3/4 inch x 1/4 inch, having a length of approximately 1 inch. Three such pieces were then plied to give a 108-ply laminate 1 inch in length and measuring approximately 3/4 inch by 3/4 inch.

The sample, frozen to stiffen the resin and stabilize the structure for cutting, was cut with a diamond saw producing film pieces varying in thickness from about 0.0125 cm to about 0.25 cm.

Film wafers with approximate dimensions of 3/4 inch by 3/4 inch and varying thicknesses were used to prepare test specimens by placing the wafers between two 5-mil copper foils and pressing. The films were then cured by heating under pressure at a temperature of approximately 200° F for two hours. On cooling, the foils of the resulting copper foil-adhesive sandwich specimens were found to be well adhered; specimen thicknesses were in the range of from 0.0125 to 0.15 cm.

Thermal conductivity of the carbon filled adhesive structures was determined both for the through-thickness and in-plane directions. Debonded samples taken from copper foil sandwich

test specimens were used for measurement of the specific heat, density and thermal diffusivity. The test method for determining thermal diffusivity by the laser flash method is set forth in ASTM E1461. Generally, the entire 0.5 inch diameter specimen disc is illuminated by laser beam, and the temperature rise on the opposing face is measured as a function of time. The rate of change, in  $\text{cm}^2/\text{sec}$ , is diffusivity; conductivity is the product of diffusivity, heat capacity and density.

The thermal conductivity of an uncured sample prepared as in Example 2 and having a thickness of 0.125 cm was determined to be 435 W/mK. The theoretical conductivity, based on the fiber volume of the adhesive film, is 500 W/mK.

- 10 Example 3 Braided, pitch-based carbon fiber tow having a thermal conductivity of 500 W/mK in the fiber axis direction, obtained as EWB-500X Thornel carbon fiber from Amoco Performance Products, was impregnated with a pre-mixed, 15 Durometer hardness, two-part thermoset silicone elastomer formulation, obtained as R-2602 silicone from NuSil Technologies. The impregnated tow strips were laid up, restrained for curing by placing in a shrink tubing mold, then cured by placing in a vacuum oven preheated at 155° C and holding for 30 min., cooling to room temperature and holding for 23.5 hr. before cutting. The resulting soft, fully cured, carbon fiber-filled elastomer had a fiber volume of 13.8 vol% (21.5 wt%). Slicing transversely to the fiber axis using a microtome provided film discs with a diameter of approximately 0.5 inches and 2.03 mm thickness, with carbon fiber extending through the thickness of the film. The thermal conductivity of the film slices was determined as 70 W/mK by the laser flash method.

Thermal resistance may be measured by the method described by J. R. Mason in "Thermal Evaluation of Simulated Finish and Flatness Conditions Typical to MCM and MPV Applications", Proceedings, IEEE SEMITHERM XII Conference, March 1996, Austin, TX.

- 25 The thermal resistance of a 0.86 mm thick film placed between aluminum blocks, was determined by this method as  $0.13^\circ \text{C in}^2/\text{W}$ . The value represents a combination of the inherent thermal resistance of the film and the contact resistance at each film/block interface.

- Example 4 Braided carbon fiber tow, obtained as EWB-400X Thornel carbon fiber from Amoco Performance Products, was impregnated with pre-mixed, two-part thermoset silicone elastomer formulation, 20 Durometer hardness, obtained as R-2188 silicone from NuSil
- 30

Technologies. The mixed resin was degassed at ambient temperature and at a reduced pressure of 25 in. Hg prior to impregnating the fiber tow. The impregnated tow strips were laid up, placed in an acrylic mold having a 4.5"x4.5"x1.5" cavity, deaerated by placing in a vacuum oven at room temperature and 29 in. Hg, cured by heating the vacuum oven at 100° C and holding for approximately 15 hr., then cooling to room temperature. The resulting soft, fully cured, carbon fiber-filled elastomer had a fiber volume of 18 vol% (28 wt%).

Slicing transversely to the fiber axis using a microtome provided 2.00 mm thick film pieces with carbon fiber extending through the thickness of the film. The thermal conductivity of the film slices was determined as 42 W/mK.

10 Example 5 Rigid epoxy matrix/carbon fiber films were prepared from unidirectional prepreg tape substantially as described in Example 2 and tested. The thermal conductivity of a sample having a thickness of 2.0 cm (80 mils) was determined to be 224 W/mK. The thermal conductivity of a specimen having a film thickness of 67 mils and a fiber volume of 50 vol% (34 wt%) was determined to be 62.2 W/mK.

15 It will thus be apparent that both rigid and soft thermally conductive interface materials may be prepared according to the invention, and with a very high thermal conductivity in the transverse direction. Prior art conductive interface adhesives and pads are disclosed to have thermal conductivities less than 10 W/mK, and most common interface materials have thermal conductivities less than 5 W/mK. The interface materials according to the invention are seen to have a thermal conductivity greater than 10 W/mK, and as great as approximately 500 W/mK, with even higher thermal conductivities readily attainable using high fiber volumes and compositions comprising highly conductive carbon fiber.

The invention will thus be seen to be a thermally conductive film, useful as an interface material in electronic applications. The film may be made adhesive to bond electronic components to thermal management components. The interface materials of this invention will comprise a matrix resin and a pitch-based carbon fiber, and will have a very high thermal conductivity in the through-thickness direction of greater than about 10 W/mK, preferably greater than about 50 W/mK and desirably greater than 100 W/mK, to as great as 800 to 1000 W/mK. The thermal conductivity of the interface materials of this invention is thus

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substantially greater than that of thermally-conductive film and film adhesives known in the art and currently available for these uses.

Preferably the interface material will have the form of a film and will comprise a matrix resin selected from thermoset resin and thermoplastic resin, and from about 10 to about 70 vol%,  
5 more preferably from about 30 to about 60 vol% carbon fiber. Alternatively, the formulations may be conveniently described as comprising from about 15 to about 80 wt%, preferably from about 15 to about 70 wt%, still more preferably from about 25 to about 70 wt% carbon fiber, based on combined weight of fiber and resin. The carbon fiber will be embedded in the film and aligned to extend substantially through the film in the thickness direction. More preferably  
10 the fiber will be aligned substantially perpendicular to an axis of the film surfaces, and still more preferably, the fiber will extend through the film and terminate at opposing faces of the film, thereby providing an efficient thermal pathway between the components adhered thereto. It will be understood that highly thermally conductive carbon fiber is also electrically conductive. Adhesive film comprising carbon fiber according to this invention may therefore  
15 also find application in bonding the conducting elements of microelectronic components, for example, conductors and contactors employed in connectors and the like.

It will be recognized by those skilled in the art that further modifications, particularly in the processes described for making the film and film adhesives of this invention, may be made without departing from the spirit and scope of the invention, which is solely defined by the  
20 appended claims.

## Claims:

1. A film having a transverse thermal conductivity greater than about 10 W/mK, said film comprising a matrix resin and from about 10 to about 70 wt% carbon fiber, said fiber extending substantially through said film in the thickness direction.
- 5 2. The film of Claim 1 wherein said film has a thickness of from 0.005 cm to 1 cm.
3. The film of Claim 1 wherein said transverse thermal conductivity is from 50 to 1500 W/mK.
4. The film of Claim 1 wherein said fiber extends through the thickness of said film, terminating at the opposing faces thereof.
- 10 5. The film of Claim 1 wherein said fiber is aligned substantially perpendicular to the plane of said film.
6. The film of Claim 1 wherein less than 50 volume % of said fiber is aligned substantially along the horizontal plane of said film.
7. The use of a film according to any of Claims 1-6 in an electronic device to fill the gap  
15 formed at the interface between an electronic component and a thermal management component.
8. The use of a film according to Claim 7 wherein said film is adhesively bonds said components.
9. A device comprising a thermal management component, at least one electronic  
20 component associated therewith, and a film according to any of Claims 1-6 interposed at the interface between said components.



# INTERNATIONAL SEARCH REPORT

Int. l. Application No

PCT/US 98/04720

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C08J5/18 C08K7/06 H01L23/373 C09J201/00

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08J C08K H01L C09J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	PATENT ABSTRACTS OF JAPAN vol. 095, no. 001, 28 February 1995 & JP 06 299129 A (NEC CORP), 25 October 1994, see abstract	1-9
Y	WO 86 00629 A (UNIV LOUVAIN ;ISSI JEAN PAUL (BE); ALLEN GORDON DEREK (BE)) 30 January 1986 see the whole document	1-9
Y	DATABASE WPI Section Ch, Week 8628 Derwent Publications Ltd., London, GB; Class A85, AN 86-180501 XP002069372 & JP 61 114 841 A (FUJIYOSHI K) see abstract	1-6

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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

### \* Special categories of cited documents:

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- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"A" document member of the same patent family

Date of the actual completion of the international search

25 June 1998

Date of mailing of the international search report

09/07/1998

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# INTERNATIONAL SEARCH REPORT

Int .tional Application No

PCT/US 98/04720

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>EP 0 456 428 A (E.I. DU PONT DE NEMOURS AND COMPANY) 13 November 1991 see the whole document &amp; US 5 026 748 A cited in the application -----</p>	1-9

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 98/04720

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 8600629 A	30-01-1986	LU 85462 A EP 0191026 A	12-02-1986 20-08-1986
EP 0456428 A	13-11-1991	US 5026748 A AT 128168 T AU 629157 B AU 7613791 A CA 2041807 A CN 1057282 A DE 69113108 D DE 69113108 T ES 2078443 T IL 97957 A JP 6212137 A MX 172824 B RU 2052483 C TR 25806 A	25-06-1991 15-10-1995 24-09-1992 07-11-1991 08-11-1991 25-12-1991 26-10-1995 09-05-1996 16-12-1995 12-04-1994 02-08-1994 14-01-1994 20-01-1996 01-09-1993